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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of
Hirokazu TANAKA, et al.

Group Art Unit: 1773
Examiner: AHMED SHEEBA

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For: coating liquid for forming Hard Coat Film and Substrate Coated
With Such a Film

Honorable Commissioner of Patents and Trademarks
United States Patent and Trademark Office
Washington, D. C. 20231

DECLARATION UNDER 37 CFR 1.132

I, Hirokazu TANAKA, declare and state that:

1. I am a citizen of Japan, and residing at 13-2, Kitaminato-machi,
Wakamatsu-ku, Kitakyushu-shi, Fukuoka, Japan.

In March 1969, I was graduated from Tokyo University of Science,
and received a degree of Bachelor of Science.

Since April 1969, I have been an employee of Catalyst &
Chemicals Industries Co., Ltd. Simultaneously, I had been
assigned to Wakamatsu Plant and engaged in the development of
novel inorganic materials, especially the production of the fine
inorganic powders.

2. I am a co-inventor of the invention described in the

specification of the above-identified application.

3. The following Experimental Test was carried out in order to demonstrate the unobviousness of the present invention.

Experiment

New Comparative Example C

0.03 part by weight, in term of Fe_2O_3 , of ferric chloride and 99.97 parts by weight, in term s of TiO_2 , of titanium tetrachloride were dissolved in pure water, thereby obtaining 10,000 parts by weight of an aqueous solution of mixture. 15% aqueous ammonia was slowly added to the aqueous solution of mixture until the pH thereof became 9.0, thereby obtaining a co-precipitate gel of iron oxide hydrate and titanium oxide hydrate.

The thus obtained co-precipitate gel was dehydrated and washed. 1150 part by weight of 35% hydrogen peroxide and 250 parts of the pure water were added to 1100 parts by weight of the co-precipitated gel, and heated to 80°C . As are result, a reddish-brown solution was obtained. This solution was diluted with pure water so that the concentration of iron and titanium oxides contained in the solution, in terms of $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$, became 1.0% by weight, and heated at 200°C for 9 hr in autoclave.

Thereafter, the water of the solution was replaced by methanol, and the mixture was concentrated until the concentration in terms of $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$ became 20% by weight. Thus there was obtained a sol of composite metal oxide of iron oxide and titanium oxide (sol F_1) having an average particle size of 11 nm and a weight ratio $\text{Fe}_2\text{O}_3/\text{TiO}_2$ of 3/9997 (0.0003).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol F₁ was used in place of the sol A₁. Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A.

New Example A

0.05 part by weight, in term of Fe₂O₃, of ferric chloride and 99.95 parts by weight, in term s of TiO₂, of titanium tetrachloride were dissolved in pure water, thereby obtaining 10,000 parts by weight of an aqueous solution of mixture. 15% aqueous ammonia was slowly added to the aqueous solution of mixture until the pH thereof became 9.0, thereby obtaining a co-precipitate gel of iron oxide hydrate and titanium oxide hydrate.

The thus obtained co-precipitate gel was dehydrated and washed. 1150 part by weight of 35% hydrogen peroxide and 250 parts of the pure water were added to 1100 parts by weight of the co-precipitated gel, and heated to 80°C. As are result, a reddish-brown solution was obtained. This solution was diluted with pure water so that the concentration of iron and titanium oxides contained in the solution, in terms of (TiO₂+Fe₂O₃), became 1.0% by weight, and heated at 200°C for 9 hr in autoclave.

Thereafter, the water of the solution was replaced by methanol, and the mixture was concentrated until the concentration in terms of (TiO₂+Fe₂O₃) became 20% by weight. Thus there was obtained a sol of composite metal oxide of iron oxide and titanium oxide

(sol G₁) having an average particle size of 11 nm and a weight ratio Fe₂O₃/TiO₂ of 5/9995 (0.0005).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol G₁ was used in place of the sol A₁. Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A.

New Example B

0.48 part by weight, in term of Fe₂O₃, of ferric chloride and 99.52 parts by weight, in term s of TiO₂, of titanium tetrachloride were dissolved in pure water, thereby obtaining 10,000 parts by weight of an aqueous solution of mixture. 15% aqueous ammonia was slowly added to the aqueous solution of mixture until the pH thereof became 9.0, thereby obtaining a co-precipitate gel of iron oxide hydrate and titanium oxide hydrate.

The thus obtained co-precipitate gel was dehydrated and washed. 1150 part by weight of 35% hydrogen peroxide and 250 parts of the pure water were added to 1100 parts by weight of the co-precipitated gel, and heated to 80°C. As are result, a reddish-brown solution was obtained. This solution was diluted with pure water so that the concentration of iron and titanium oxides contained in the solution, in terms of (TiO₂+Fe₂O₃), became 1.0% by weight, and heated at 200°C for 9 hr in autoclave.

Thereafter, the water of the solution was replaced by methanol, and the mixture was concentrated until the concentration in terms

of $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$ became 20% by weight. Thus there was obtained a sol of composite metal oxide of iron oxide and titanium oxide (sol H_1) having an average particle size of 11 nm and a weight ratio $\text{Fe}_2\text{O}_3/\text{TiO}_2$ of 48/9952 (**0.0048**).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol H_1 was used in place of the sol A_1 . Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A.

New Example C

0.15 part by weight, in term of Fe_2O_3 , of ferric chloride and 99.85 parts by weight, in term s of TiO_2 , of titanium tetrachloride were dissolved in pure water, thereby obtaining 10,000 parts by weight of an aqueous solution of mixture. 15% aqueous ammonia was slowly added to the aqueous solution of mixture until the pH thereof became 9.0, thereby obtaining a co-precipitate gel of iron oxide hydrate and titanium oxide hydrate.

The thus obtained co-precipitate gel was dehydrated and washed. 1150 part by weight of 35% hydrogen peroxide and 250 parts of the pure water were added to 1100 parts by weight of the co-precipitated gel, and heated to 80°C. As are result, a reddish-brown solution was obtained. This solution was diluted with pure water so that the concentration of iron and titanium oxides contained in the solution, in terms of $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$, became 1.0% by weight, and heated at 200°C for 9 hr in autoclave.

Thereafter, the water of the solution was replaced by methanol, and the mixture was concentrated until the concentration in terms of $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$ became 20% by weight. Thus there was obtained a sol of composite metal oxide of iron oxide and titanium oxide (sol I_1) having an average particle size of 11 nm and a weight ratio $\text{Fe}_2\text{O}_3/\text{TiO}_2$ of 15/9985 (**0.0015**).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol I_1 was used in place of the sol A_1 . Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A.

TableA

	sol		Photo chromism	High refractivity	Scuffing Resistance	Appearance	Dye affinity	water resistance		Cloudiness	Stability	
	Fe ₂ O ₃ /TiO ₂	SiO ₂ / (Fe ₂ O ₃ +TiO ₂)						coloring	adherence		25 days	45 days
Comp. Ex. C(new)	3/9997		exhibited (blue)	0	B	0	0	occured	0	0	0	x
Ex. A(new)	5/9995		none	0	A	0	0	none	0	0	0	Δ
Ex. B(new)	48/9952		none	0	A	0	0	none	0	0	0	Δ
Ex. C(new)	15/9985		none	0	A	0	0	none	0	0	0	Δ

4. From the result of the above Experiment and based on my best knowledge and experience on the inorganic material development,

I conclude as follows.

The composite metal oxide having the limited weight ratio (0.0005 to 0.005) of iron oxide to titanium oxide of the present invention is free from photochromism. Further, the obtained hard coat film is excellent in weather resistance, and has a high surface hardness to thereby ensure excellent scuffing resistance and attrition resistance. The hard coat film does not have photochromism, so that it is free from discoloration by light irradiation.

Accordingly, the composite metal oxide having the limited weight ratio of the iron oxide to the titanium oxide of the present invention is specifically free from photochromism.

The undersigned declares further that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This *15* day of May 2003


Hirokazu TANAKA